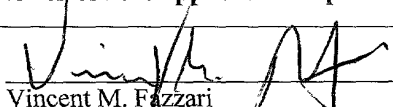


FORM PTO-1390 (REV 10-94)		U.S. DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE		DOCKET #: 4080-38PUS
TRANSMITTAL LETTER TO THE UNITED STATES DESIGNATED/ELECTED OFFICE (DO/EO/US) CONCERNING A FILING UNDER 35 U.S.C. 371				
				U.S. APPLICATION NO. (If known, see 37 CFR 1.5) <b>09/913008</b>
INTERNATIONAL APPLICATION NO. <b>PCT/DE00/00384</b>		INTERNATIONAL FILING DATE <b>February 10, 2000</b>		PRIORITY DATE CLAIMED <b>February 11, 1999</b>
TITLE OF INVENTION <b>Multicomponent Coating and Adhesive Material</b>				
APPLICANT(S) FOR DO/EO/US <b>Christian TERFLOTH; Theodor HIPOLD</b>				
Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:				
<p>1. <input checked="" type="checkbox"/> This is a <b>FIRST</b> submission of items concerning a filing under 35 U.S.C. 371.</p> <p>2. <input type="checkbox"/> This is a <b>SECOND</b> or <b>SUBSEQUENT</b> submission of items concerning a filing under 35 U.S.C. 371</p> <p>3. <input checked="" type="checkbox"/> This express request to begin national examination procedures (35 U.S.C. 371(f)) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U.S.C. 371(b) and PCT Articles 22 and 39(1).</p> <p>4. <input checked="" type="checkbox"/> A proper Demand for International Preliminary Examination was made by the 19th month from the earliest claimed priority date.</p> <p>5. <input checked="" type="checkbox"/> A copy of the International Application as filed (35 U.S.C. 371(c)(2))</p> <p>6. <input checked="" type="checkbox"/> A translation of the International Application into English (35 U.S.C. 371(c)(2)).</p> <p>7. <input type="checkbox"/> Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3))</p> <p>8. <input type="checkbox"/> A translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).</p> <p>9. <input checked="" type="checkbox"/> An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)).</p> <p>10. <input type="checkbox"/> A translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)).</p> <p><b>Items 11. to 16. Below concern other document(s) or information included:</b></p> <p>11. <input checked="" type="checkbox"/> An Information Disclosure Statement under 37 CFR 1.97 and 1.98.</p> <p>12. <input type="checkbox"/> An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.</p> <p>13. <input checked="" type="checkbox"/> A <b>FIRST</b> preliminary amendment. <input type="checkbox"/> A <b>SECOND</b> or <b>SUBSEQUENT</b> preliminary amendment.</p> <p>14. <input type="checkbox"/> A substitute specification.</p> <p>15. <input type="checkbox"/> A change of power of attorney and/or address letter.</p> <p>16. <input checked="" type="checkbox"/> Other items or information (<i>specify</i>): PCT Publication Sheet, Int'l Preliminary Examination Report, Int'l Search Report, PCT Request</p>				

U.S. APPLICATION NO. (if known, see 37 CFR 1.5) <b>09/913008</b>		INTERNATIONAL APPLICATION NO <b>PCT/DE00/00384</b>		ATTORNEY'S DOCKET NUMBER <b>4080-38PUS</b>	
17.[x]The following fees are submitted:					
<b>Basic National Fee (37 CFR 1.492(a)(1)-(5)):</b>					
Search Report has been prepared by the EPO or JPO .....				<b>\$860.00</b>	
International preliminary examination fee paid to USPTO (37 CFR 1.482).....				<b>\$690.00</b>	
No international preliminary examination fee paid to USPTO (37 CFR 1.482) but international search fee paid to USPTO (37 CFR 1.445(a)(2)) .....				<b>\$710.00</b>	
Neither international preliminary examination fee (37 CFR 1.482) nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO .....				<b>\$1000.00</b>	
International preliminary examination fee paid to USPTO (37 CFR 1.482) and all claims satisfied provisions of PCT Article 33(2)-(4) .....				<b>\$100.00</b>	
<b>ENTER APPROPRIATE BASIC FEE AMOUNT =</b>				<b>\$860.00</b>	
Surcharge of <b>\$130.00</b> for furnishing the oath or declaration later than <input type="checkbox"/> 20 <input type="checkbox"/> 30 months from the earliest claimed priority date (37 CFR 1.492(e)).				\$	
Claims	Number Filed	Number Extra	Rate		
Total Claims	29 - 20 =	9	x <b>\$18.00</b>	<b>\$162.00</b>	
Independent Claims	4 - 3 =	1	x <b>\$80.00</b>	<b>\$80.00</b>	
Multiple dependent claim(s) (if applicable)			+ <b>\$270.00</b>	\$	
<b>TOTAL OF ABOVE CALCULATIONS =</b>				<b>\$1102.00</b>	
Reduction of 1/2 for filing by small entity, if applicable.				\$	
<b>SUBTOTAL =</b>				<b>\$1102.00</b>	
Processing fee of <b>\$130.00</b> for furnishing the English translation later than <input type="checkbox"/> 20 <input type="checkbox"/> 30 months from the earliest claimed priority date (37 CFR 1.492(f)).				\$	
<b>TOTAL NATIONAL FEE =</b>				<b>\$1102.00</b>	
Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by the appropriate cover sheet (37 CFR 3.28, 3.31). <b>\$40.00</b> per property				\$	
<b>TOTAL FEES ENCLOSED</b>				\$	
Amount to be refunded:				\$	
Charged:				\$	
<p>a. [x] One check in the amount of \$1102.00 to cover the above fees is/are enclosed.</p> <p>b. <input type="checkbox"/> Please charge my Deposit Account No. <u>03-2412</u> in the amount of \$_____ to cover the above fees. A duplicate copy of this sheet is enclosed.</p> <p>c. [x] The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any overpayment to Deposit Account No. <u>03-2412</u>. A duplicate copy of this sheet is enclosed.</p> <p><b>NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137(a) or (b)) must be filed and granted to restore the application to pending status.</b></p>					
SEND ALL CORRESPONDENCE TO: <u>Vincent M. Fazzari</u> Cohen, Pontani, Lieberman & Pavane 551 Fifth Avenue, Suite 1210 New York, New York 10176			 <u>Vincent M. Fazzari</u> Registration Number: <u>26,879</u> Tel: (212) 687-2770		

Attorney Docket # 4080-38PUS

Patent

**IN THE UNITED STATES PATENT AND TRADEMARK OFFICE**

In re National Phase PCT Application of  
Christian TERFLOTH et al.  
International Application No. PCT/DE00/00384  
International Filing Date: February 10, 2000  
For: Multicomponent Coating and Adhesive Material

Assistant Commissioner for Patents  
Washington, DC 20231  
**BOX PCT**

**PRELIMINARY AMENDMENT**

S I R:

Please preliminarily amend the above-identified application as follows:

**IN THE SPECIFICATION:**

Page 1, between title and first line insert:

**--Background of the Invention**

Reactive, solvent-free adhesives cross-linkable with moisture from the air, particularly hot-melt adhesives, are well known in the state of the art. Such adhesives are prepared from low-molecular starting materials comprising OH groups, such as polyesters or polyethers, which are then converted to reactive isocyanate-terminated adhesives with a stoichiometric excess of isocyanates. 4,4'-diisocyanato-diphenylmethane (MDI), 1,5-diisocyanato-naphthalene (NDI), 1,6-diisocyanato-hexane (HDI), 2,4-diisocyanato-toluene (TDI), 1-isocyanato-3-isocyanatomethyl-3,5,5-trimethylcyclohexane (IPDI) or their prepolymers can be used as isocyanate components.--.

Page 2, at line 29, insert:

**--The Invention**

The afore-mentioned technical problems are solved by the subject matter of the invention. In one aspect of the invention, it is directed to a continuous preparation of the inventive isocyanate-terminated coating and adhesive material (e.g. just at the site where its application is desired) by mixing the components and by heating them, particularly under continued mixing, until a liquid state is reached, in which the material is then available as a reactive coating and adhesive material.--.

Page 4, at line 6 insert:

--In the silane-functionalized coating and adhesive material the first component comprises a higher-molecular starting polymer and a second component comprises a reactive, silane-functionalized cross-linking agent.--.

Page 10, following the last line insert as a separate paragraph --The terms and expressions which have been employed are used as terms of description and not of limitation, and there is no intention in the use of such terms and expressions of excluding any equivalents of the features shown and described or portions thereof, it being recognized that various modifications are possible within the scope of the invention--.

**IN THE CLAIMS:**

Without prejudice, cancel claims 1-17 and add claims 18-46 as follows:

--18. A reactive isocyanate-terminated multicomponent coating and/or adhesive material comprising:

- (a) a first component comprising an isocyanate-reactive polymer having a molecular weight of more than 8,000 g/mol;
- (b) a second component comprising a reactive isocyanate-terminated cross-linking agent; and
- (c) a polymer which is not isocyanate-reactive and which is selected from the group consisting of an ethylene/vinyl-acetate copolymer and/or a polyolefin.

19. The multicomponent coating and/or adhesive material of claim 18 wherein the ethylene/vinylacetate copolymer has a vinyl-acetate content of from 12 to 40%, particularly 18 to 28%, and a melting indice of from 8 to 800, particularly 150 to 500.

20. The multicomponent coating and/or adhesive material of claim 18, wherein the polyolefin has an average molecular weight  $M_n$  of from 5,000 to 25,000 g/mol, particularly 10,000 to 20,000 g/mol, and a softening range of from 80° to 170°C, particularly 80° to 130°C.

21. The multicomponent coating and/or adhesive material of claim 18, wherein the first component contains 5 to 35 wt.% of said non-isocyanate-reactive polymer.

22. The multicomponent coating and/or adhesive material of claim 18 wherein, the reactive cross-linking agent comprises an isocyanate solid at room temperature, particularly wherein said isocyanate solid at room temperature is selected from the group consisting of 4,4'-

diisocyanato-diphenylmethane (MDI), 4,4',4''-triisocyanato-triphenylmethane, tris-(4-isocyanatophenyl)-thiophosphate, I,5-diisocyanato-naphthalene (N DI) and/or isomers thereof, dimers (dimerisates) of 2,4-diisocyanato-toluene (TDI) and/or of 1-isocyanato-3-isocyanatomethyl-3,5,5-trimethylcyclohexane (I PDI) and/or their hydration products and/or trimers (trimerisates) of 1-isocyanato-3-isocyanatomethyl-3,5,5-trimethylcyclohexane (IPDI).

23. The multicomponent coating and/or adhesive material of claim 18, wherein the isocyanate-reactive starting polymer has an average molecular weight of from 8000 to 50,000 g/mol, particularly between 10,000 to 30,000 g/mol, wherein said isocyanate-reactive starting polymer is selected from the group consisting of polyesters, polycaprolactonepolyesters, polyethers, polyurethanes, polyamides and polytetrahydrofuranes and/or has at least two reactive groups with reactive hydrogen atoms per molecule.

24. The multicomponent coating and/or adhesive material of claim 18, wherein the content of said isocyanate-reactive polymer is 20 to 100 wt.%, particularly 50 to 95 wt.%.

25. The multicomponent coating and/or adhesive material of claim 18, wherein at least one of (a), (b) and (c) is present as a granulate (granular material).

26. The multicomponent coating and/or adhesive material of claim 18, wherein at least one of (a), (b) and (c) comprise at least one resin, wherein said resin is selected from the group consisting of aliphatic, cyclic or cycloaliphatic hydrocarbon resins, terpene phenol resins, cumarone-indene resins,  $\alpha$ -methyl styrene resins, polymerized tall resin esters and ketone aldehyde resins and/or particularly wherein said resin is a resin having a low acid number with

values less than 1 mg KOH/g and/or particularly wherein the respective content (amount) of said resin in one or more components is from 0 to 70 wt.%, preferably 5 to 35 wt.%.

27. The multicomponent coating and/or adhesive material of claim 18, wherein the mixing ratio (blending ratio) of (a) to (b) is from 20:1 to 1:20.

28. The multicomponent coating and/or adhesive material of claim 18, wherein at least one of said (a), (b) and (c) comprises at least one further additive known per se with respect to reactive adhesives, particularly softeners (plasticizers) such as softeners (plasticizers) based on phthalic acids or phosphoric acid esters, glycol acetate, high-boiling organic oils, esters or other additives inducing plastification, stabilizers, antioxidant agents (corrosion inhibitors), acid trappers (acidtrapping agents), and/or age inhibitors.

29. A reactive isocyanate-terminated multicomponent coating and/or adhesive material comprising: an isocyanate-reactive starting polymer having a molar mass of more than 8,000 g/mol, a reactive isocyanate-terminated cross-linking agent; and a polymer which is not isocyanate-reactive and selected from the group consisting of an ethylene/vinylacetate copolymer and/or a polyolefin.

30. The multicomponent coating and/or adhesive material of Claim 29, wherein said material is moisture-reactive.

31. A method for preparing a reactive isocyanate-terminated multicomponent coating

and/or adhesive material comprising:

(a) mixing or blending a first and a second component wherein:

(i) the first component comprises an isocyanate-reactive starting polymer having a molecular weight of at least 8,000 g/mol and

(ii) the second component comprises a reactive isocyanate-terminated cross-linking agent and

wherein said multicomponent coating and/or adhesive material further comprises a polymer which is not isocyanate-reactive and which is selected from the group consisting of an ethylene/vinylacetate copolymer and/or a polyolefin; and

(b) heating the components while mixing or blending to a liquid state.

32. The method of claim 31 wherein the resulting reactive multicomponent coating and/or adhesive material, immediately after being prepared, is fed or conveyed, optionally via intermediate containers (buffer tanks), to a profile sheathing plant or to a coating plant for use.

33. A method for preparing a reactive isocyanate-terminated multicomponent coating and/or adhesive material comprising:

(a) mixing or blending a first and a second component wherein:

(i) the first component comprises an isocyanate-reactive starting polymer having a molecular weight of at least 8,000 g/mol and

(ii) the second component comprises a reactive isocyanate-terminated cross-linking agent and wherein said multicomponent coating and/or adhesive material further



comprises a polymer which is not isocyanate-reactive and which is selected from the group consisting of an ethylene/vinylacetate copolymer and/or a polyolefin, and that the resulting reactive multicomponent coating and/or adhesive material, immediately after being prepared, is applied, optionally via intermediate containers (buffer tanks), by spraying, by injection, by nozzle (die) application or by roller application (roller coating)

34. The method of claim 31 wherein the content of said non-isocyanate-reactive polymer in said first component preferably is in the range of from 5 to 35 wt%.

35. The method of claim 31, wherein said ethylene/vinylacetate copolymer has a vinylacetate content of from 12 to 40 %, particularly 18 to 28 %, and a melting indice of from between 8 to 800, particularly 150 to 500.

36. The method of claim 31, wherein said polyolefin has an average molecular weight  $M_n$  of from 5,000 to 25,000 g/mol, preferably 10,000 and 20.000 g/mol and a softening range of from 80 to 170 °C, preferably 80° to 130 °C.

37. The method any of claim 31 wherein said reactive cross-linking component comprises an isocyanate which is solid at room temperature and/or preferably selected from the group consisting of 4,4'-diisocyanato-diphenylmethane (MDI), 4,4',4''-triisocyanato-triphenylmethane, tris-(4-isocyanatophenyl)-thiophosphate, 1, 5-di isocyanato-naphthalene (NDI) and/or isomers thereof, dimers (dimerisates) of 2,4-diisocyanato-toluene (TDI) and/or of 1-isocyanato-3-isocyanatomethyl-3,5,5-trimethylcyclohexane (IPDI) and/or their hydration

products and/or trimers (trimerisates) of 1-isocyanato-3-isocyanatomethyl-3,5,5-trimethyl-cyclohexane (IPDI).

38. The method of claim 31 wherein said isocyanate-reactive starting polymer has an average molecular weight of from 8,000 to 50,000 g/mol, preferably 10,000 to 30,000 g/mol, particularly wherein said isocyanate-reactive starting polymer is preferably selected from the group consisting of polyesters, polycaprolactonepolyesters, polyethers, polyurethanes, polyamides and polytetrahydrofuranes and/or preferably has at least two reactive groups with reactive hydrogen atoms per molecule.

39. The method of claim 31 wherein said first component has a content of said isocyanate-reactive starting polymer from 20 to 100 wt.%, preferably 50 to 95 wt.%.

40. The method of claim 31 wherein the at least one adhesive component is a granulate (granular material).

41. The method of claim 31 wherein at least one or more of the components has at least one resin, particularly wherein said resin is preferably selected from the group consisting of aliphatic, cyclic or cycloaliphatic hydrocarbon resins, terpene phenol resins, cumarone-indene resins,  $\alpha$ -methyl styrene resins, polymerized tall resin esters and/or the ketone aldehyde resins and/or particularly wherein said resin is preferably a resin with a low acid number with values lower than 1 mg KOH/g and/or particularly wherein the respective amount of said resin in said least one component is 0 to 70 wt.%, preferably 5 to 35 wt.%.

42. The method of claim 31 wherein said first and said second components have a mixing ratio (blending ratio) of from 20:1 and 1:20.

43. A method of bonding materials or continuously coating materials comprising applying the multicomponent coating and/or adhesive material of claim 18 to the material to be bonded or continuously coated.

44. The method of claim 43 wherein the applying is by spraying, by injection, by nozzle (die) application or by roller application (roller coating).

45. The method of claim 43 wherein the materials are bonded and said materials are foam materials and cushions, upholstered furniture and mattresses,

46. The method of claim 43 wherein the continuous coating is for profile sheathing or cladding.

IN THE ABSTRACT:

--Disclosed is a multicomponent, isocyanate-terminated or silane-functionalized coating and adhesive material which is continuously produced by mixing the individual components and by heating the latter and continuing mixing until said components reach a liquid state, wherein the resulting reactive coating and adhesive material is fed for direct use especially in profile sheathing plants, coating plants or the like. In the case of isocyanate-terminated material, one component has a reactive high-molecular weight starting polymer and a second

component has a reactive isocyanate-terminated cross-linking agent. In the case of the silane-functionalized material, one component has a high-molecular weight starting material and a second component has a reactive, silane-functionalized cross-linking agent.--.

**REMARKS**

The specification, claims and abstract of the above-identified application have been amended to a form more consistent with U.S. practice.

Early consideration and allowance of the application with claims 18 to 46 are earnestly solicited

Respectfully submitted,

COHEN, PONTANI, LIEBERMAN & PAVANE

By

  
\_\_\_\_\_  
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August 7, 2001

## Version with Markings to Show Changes

### In the Specification:

Page 2, between the title and first line of text insert as a centered heading

### **--Background of the Invention--;**

Page 3, at line 29, insert as a centered heading **--The Invention--;**

line 31, delete "Claims 1...Claim 1" and insert therefore --the invention.

In one aspect of the invention, it--;

Page 4, line 6 delete "according to Claim 2,";

Page 10, following the last line, insert as a separate paragraph --The terms and expressions which have been employed are used as terms of description and not of limitation, and there is no intention in the use of such terms and expressions of excluding any equivalents of the features shown and described or portions thereof, it being recognized that various modifications are possible within the scope of the invention--.

### In the Abstract:

[The present invention relates to] Disclosed is a multicomponent, isocyanate-terminated or silane-functionalized coating and adhesive material which is continuously produced by mixing the individual components and by heating the latter and continuing mixing until said components reach a liquid state, wherein the resulting reactive coating and adhesive material is fed for direct use especially in profile sheathing plants, coating plants or the like. In the case of isocyanateterminated material, one component has a reactive high-molecular weight starting polymer and a second component has a reactive isocyanate-terminated cross-linking agent. In

the case of the silane-functionalized material, one component has a high-molecular weight starting material and a second component has a reactive, silane-functionalized cross-linking agent.

2025-04-10 10:10:10

### Multicomponent Coating and Adhesive Material

Reactive, solvent-free adhesives cross-linkable with moisture from the air, particularly hot-melt adhesives, are well known in the state of the art. Such adhesives are prepared from low-molecular starting materials comprising OH groups, such as polyesters or polyethers, which are then converted to reactive isocyanate-terminated adhesives with a stoichiometric excess of isocyanates. 4,4'-diisocyanato-diphenylmethane (MDI), 1,5-diisocyanato-naphthalene (NDI), 1,6-diisocyanato-hexane (HDI), 2,4-diisocyanato-toluene (TDI), 1-isocyanato-3-isocyanatomethyl-3,5,5-trimethylcyclohexane (IPDI) or their prepolymers can be used as isocyanate components.

Alternatively, said reactive coating and adhesive materials can also be produced on the basis of a silane-functionalized polymer. The addition of catalysts, resins and other additives and adjuvants is usual.

Depending on the above starting materials and their average molecular weights and on the desired properties of the adhesive to be produced, the underlying polyaddition reaction can take several hours. Two or more adhesive components are brought together and are processed, under the effect of heat, to give a hot, liquid mass which, in this state, can be used as a hot-melt adhesive. The cross-linking reaction takes place by taking moisture from the air and/or from a wetted surface. This means that after their preparation, these hot-melt adhesive must be kept dry, with any moisture from the air being eliminated.

Therefore, for transportation and storage such an adhesive is filled into tightly sealable containers (e.g. 20-liter or 200-liter tin-barrels) and is stored in a dry state there, with any moisture from the air being eliminated. Before use, the cooled and generally solid adhesive must be melted out of these containers and conveyed or fed to the processing and application units. Melting is achieved, for example, by means of a barrel-melter (melting device) whose heatable piston provided with sealing rings or joints is brought under pressure into a container filled with said adhesive. By means of a gear-type pump installed in the piston, the molten and viscous to fluid hot-melt adhesive is conveyed or fed through heated conduits or pipes to an application unit. Depending on the size of the

container and on the adhesive formulation, the melting capacity of such a barrel-melter is in the range of about from 20 to 80 kg/hour.

5 In some fields of application such a melting capacity is, however, not sufficient, particularly not for the coating of flat surfaces. In addition, when relatively small barrels are used, it is impossible to achieve substantially continuous operation because of frequent changing of said barrels. Besides, the purchase of a barrel melter by the user means a considerable capital investment. If the container which contains the hot-melt adhesive becomes leaky during transportation, the  
10 adhesive can rapidly become unusable, particularly if it is exposed to moisture. Another disadvantage is that after opening the container, the adhesive must be processed completely and in a relatively short time because any adhesive residues become unusable relatively rapidly.

15 Another disadvantage of the state of the art described before is the restriction that the use of such systems is limited exclusively to reactive adhesives only. The alternating processing of reactive adhesives and thermoplastic adhesives (based on EVA, APAO or PA) is disadvantageous for both economical and technical reasons.

20 The use of multicomponent systems in the production of hot-melt adhesives and coating materials is generally known per se. A feature common to the systems disclosed in EP 0 304 026 B1, in the Handbook of Adhesives, 2nd Edition, p. 581 ff and in the ICI polyurethanes book, 2nd Edition, p. 93 ff, is that the components exclusively react with one another and that, after the reaction has taken  
25 place, they no longer have any reactivity for secondary reactions, e.g. with moisture. Also, the components have necessarily to be used and consumed directly after mixing.

30 This afore-mentioned technical problems are solved by the subject-matter of Claims 1 and 2, wherein Claim 1 is directed to a continuous preparation of the inventive isocyanate-terminated coating and adhesive material (e.g. just at the site where its application is desired) by mixing the components and by heating them, particularly under continued mixing, until a liquid state is reached, in  
35 which the material is then available as a reactive coating and adhesive material.



For the isocyanate-terminated coating and adhesive material, it is further provided that at least one component has at least one higher-molecular, reactive starting polymer and a second component has a reactive isocyanate-terminated cross-linking agent.

5

In the silane-functionalized coating and adhesive material according to Claim 2, the first component comprises a higher-molecular starting polymer and a second component comprises a reactive, silane-functionalized cross-linking agent.

10

In the method according to the present invention, a first and at least one second adhesive component are brought into e.g. a screw conveyor for mixing and heating these components in order to produce a two- or multicomponent hot-melt adhesive. There, the components are melted and mixed, wherein a coating and/or adhesive material is produced by applying appropriate process parameters (such as e.g. temperature, pressure, screw length etc.), said parameters depending on the desired coating or adhesive material and on the composition of the components; the resulting coating and/or adhesive material can be processed or used immediately, particularly it can be conveyed or fed immediately to its application, preferably to a coating process.

15

20

In contrast to the prior art methods where the production installations of the adhesive and the application installations on the other hand are not the same and thus transportation of said adhesive is necessary, said transportation leading to the described disadvantages, the inventive coating and adhesive material is produced in situ at the user who, for example, has available an appropriate screw conveyor, which is economically and technically advantageous. The quantity of adhesive which is actually needed can be specifically produced and this in a continuous process. The disadvantages during transportation or after opening the container according to the state of the art described above thus do not arise in the case of the present invention.

25

30

According to a first embodiment of the present invention, an isocyanate termination of the adhesive is provided, particularly one according to which the first component has at least one higher-molecular reactive starting polymer and the second component comprises an isocyanate-terminated cross-linking agent.

35

According to a second embodiment of the present invention, a silane-functionalization of the adhesive is provided, particularly one according to which the second component has a silane-functionalized cross-linking agent.

5 The first and the second adhesive component can be stored separately from each other, thus resulting in problem-free storage and transportation possibilities and prolonged storage times. According to one embodiment of the present invention, both components can occur advantageously in granular form, wherein, however, liquid storage and transportation forms are generally also possible.

10 Moreover, it is easy to achieve sufficiently high melting capacities of more than 100 kg/hour or more and to prepare large quantities of coating or adhesive material with a screw conveyor. Such a screw conveyor is known per se, for example, as a single-screw extruder.

15 All adjustments of the screw conveyor (such as temperature, pressure, throughput time etc.) are determined and adapted as a function of the coating and adhesive material to be produced and thus as a function of the compositions of the components. The pressure prevailing along the conveying track should be sufficient to heat the components, wherein, however, additional heat sources can also  
20 be provided so that temperatures of about 200 °C can easily be reached along a short conveying track.

25 An optionally heatable pipe or conduit conveying or feeding the prepared hot-melt adhesive in liquid form directly to the processing site can be connected to the screw conveyor at the end of the conveying track. In an advantageous manner, the coating or adhesive according to the present invention is produced only when there is a respective need. After using the coating or adhesive, it is easy to clean all the devices brought in contact with the inventive material inso-  
30 far as only one of the adhesive components, particularly the adhesive component free from the cross-linking agent, is passed through the screw conveyor, the corresponding pipes/conduits and other installations, thereby achieving a cleaning and flushing.

In addition, when the adhesive is prepared in relatively large quantities, it may also be advantageous to convey or feed the adhesive to the respective use via intermediate containers (intermediate storage containers, buffer tanks). Hereby, a buffer with respect to the preparation of the adhesive and/or and adaptation to given local conditions may be achieved.

According to the present invention, the first adhesive component may comprise at least one polymer which is reactive particularly with respect to isocyanates and/or a resin component and, optionally, a non-reactive polymer. The use of polymers reactive particularly with respect to isocyanates and having average molecular weights of more than 8000 g/mol has proved particularly advantageous. Such higher-molecular components, particularly having average molecular weights of 10,000 to 30,000 g/mol, e.g. polyesters, polycaprolactonepolyesters, polyethers, polyurethanes, polyamides or polytetrahydrofuranes, which have at least two reactive groups with reactive hydrogen atoms per molecule, are capable of reacting with isocyanates. The use of such reactive polymers in quantities of from 20 to 100 wt.%, particularly in quantities of from 50 to 95 wt.%, is preferred in the first adhesive component.

Resins which can be used in the first adhesive component (such as e.g. aliphatic, cyclic or cycloaliphatic hydrocarbon resins, terpene phenol resins, cumarone-indene resins,  $\alpha$ -methyl styrene resins, polymerized tall resin ester or ketone aldehyde resins) are not particularly limited. However, resins having low acid numbers, particularly having values lower than 1 mg KOH/g, are preferably used. The contents or amounts of resin(s) in the first adhesive component can, for example, preferably be about 5 to 35 wt.% and generally between 0 and 70 wt.%.

The first adhesive component may also comprise a non-reactive polymer, wherein, when using ethylene/vinylacetate copolymers, such polymers are preferred which have vinylacetate contents of from 12 to 40 %, particularly 18 to 28 %, and melting indices (DIN 53735) of from 8 to 800, particularly 150 to 500. However, polyolefins may also be used in an advantageous manner. Polyolefins having average molecular weights  $M_n$  of from 5000 to 25,000 g/mol, particularly 10,000 to 20,000 g/mol, and having softening ranges (according to the ring and

ball method) of between 80 and 170 °C, particularly 80 and 130 °C, can be used advantageously in the method according to the present invention. The amounts of the non-reactive polymer(s) in the first adhesive component are not particularly critical and are selected according to the desired coating or adhesive. For example, 5 to 35 wt.% of non-reactive polymer may be present in the first adhesive components.

The second adhesive component comprises at least one reactive cross-linking component which may be, according to the first embodiment of the present invention, at least one isocyanate solid at room temperature, or alternatively, according to the second embodiment of the present invention, a silanized polymer solid at room temperature in the case of a silane-functionalized adhesive.

The following isocyanate-terminated cross-linking agents have proven to be particularly advantageous: 4,4'-diisocyanato-diphenylmethane (MDI), 4,4',4"-triisocyanato-triphenylmethane, tris-(4-isocyanatophenyl)-thiophosphate, 1,5-diisocyanato-naphthalene (NDI) or isomers thereof, dimers (dimerisates) of 2,4-diisocyanato-toluene (TDI) and of 1-isocyanato-3-isocyanatomethyl-3,5,5-trimethyl cyclohexane (IPDI) or their hydration products and trimers (trimerisate) of 1-isocyanato-3-isocyanatomethyl-3,5,5-trimethyl cyclohexane (TPDI).

According to the present invention the adhesive components may comprise various other additives generally known for reactive adhesives, such as softeners (plasticizers), e.g. softeners based on phthalic acids or phosphoric acid esters, glycolacetate, high-boiling organic oils, esters or other additives inducing plastification, stabilizers, antioxidants (corrosion inhibitors), acid trappers (acid-trapping agents), and/or age inhibitors. These optional components or constituents of the adhesive are selected according to the field of application of the final adhesive product. Such selection is known to the person skilled in the art.

The coating and/or adhesive materials produced or prepared according to the present invention may be used, for example, for the bonding of foam materials and cushions, upholstered furniture and mattresses or can be used for continuous coating purposes, such as profile sheathing (profile cladding / profile coating / profile wrapping). Application can be made using known methods, e.g. by

spraying, by injection, by nozzle (die) application or by roller application (roller coating).

- 5 Examples for the method according to the present invention and the adhesive components to be used in said methods are given in the following.

**Example 1: Isocyanate-terminated adhesive**

5 A first adhesive component in the form of a thermoplastic granular material having the following composition was prepared:

- Polyester having an average molecular weight of about 20,000 g/mol and an OH number of 5: 60 wt.%
- Ethylene/vinylacetate copolymer having a VA content of 18 % and a melting index of about 150: 20 wt.%
- Low-molecular aromatic hydrocarbon resin having a softening range of from 75 to 85 °C and an acid number of less than 1 mg KOH/g: 20 wt.%

15 A second adhesive component was prepared in the form of a thermoplastic granular material having the following composition:

Polyisocyanate based on the cycloaliphatic 1-isocyanato-3-isocyanatomethyl-3,5,5-trimethyl-cyclohexane (IPDI) having an isocyanate content of 17 % and a functionality between 3 and 4.

25 The first adhesive component was supplied via a first feed device of a single-screw extruder heated to 200 °C to the conveying track of the extruder whilst the second adhesive component was supplied in a dosed fashion via a second feed device with a mixing ratio of 7:1. The melting capacity was 120 kg/hour.

30 An isocyanate-terminated reactive hot-melt adhesive was obtained with which it was possible to manufacture profiles consisting of MDF (Medium Density Fiberboard) or chipboard cores with veneer, decorative paper or plastic films using known profile sheathing plants (profile cladding installations).

**Example 2: Silane-functionalized adhesive**

A first adhesive component was prepared in the form of a thermoplastic granular material with the following composition:

- 31.2 wt.% polyolefin  
(Vestoplast 708, Degussa Hüls AG)
- 62.4 wt.% Hydrocarbon resin  
(Escorez 5320, Exxon Deutschland GmbH)
- 6.2 wt.% Wax  
(Parafint)
- 0.2 wt. % Dibutyl tin dilaurate catalyst  
(DBTL from Huntsmann Corp.)

A second adhesive component was prepared in the form of a thermoplastic granular material having the following composition:

- 95 wt. % silanized polyolefin  
(Vestoplast 206 V, Degussa Hüls AG)
- 5 wt.% PE wax  
(Polywachs 1000)

The first adhesive component was supplied via a first feed device of a single-screw extruder heated to 200 °C to the conveying track of the extruder whilst the second adhesive component was supplied in a dosed fashion via a second feed device with a mixing ratio of 1:1.9. The melting capacity was 120 kg/hour.

A silane-functionalized reactive hot-melt adhesive was obtained with which it was possible to manufacture profiles consisting of MDF or chipboard cores with veneer, decorative paper or plastic films using known profile sheathing plants (profile cladding installations).

**Claims:**

1. Multicomponent, isocyanate-terminated coating and adhesive material,  
**characterized by** a continuous production by mixing the individual  
components and their heating, particularly under continued mixing, until a  
liquid state is reached in which the coating and adhesive material being  
reactive then, is supplied, particularly directly, to profile sheathing plants,  
coating plants and the like for use, wherein one component has a higher-  
molecular reactive starting polymer and a second component has a reactive  
isocyanate-terminated cross-linking agent.
2. Multicomponent, silane-functionalized coating and adhesive material,  
**characterized by** a continuous production by mixing the individual  
components and their heating, particularly under continued mixing, until a  
liquid state is reached in which the coating and adhesive material being  
reactive then, is supplied, particularly directly, to profile sheathing plants,  
coating plants and the like for use, wherein one component has a higher-  
molecular starting polymer and a second component has a reactive silane-  
functionalized cross-linking agent.
3. Coating and adhesive material according to one or more of the preceding  
Claims, characterized in that at least one adhesive component is present in  
the form of a granulate (granular material).
4. Coating and adhesive material according to one or more of the preceding  
Claims, characterized in that said starting polymers being reactive particu-  
larly with respect to isocyanates have an average molecular weight between  
8000 and 50,000 g/mol, particularly between 10,000 and 30,000 g/mol.
5. Coating and adhesive material according to one or more of the preceding  
Claims, characterized in that said polymers being reactive particularly with  
respect to isocyanates are selected from the group consisting of polyesters,  
polycaprolactonepolyesters, polyethers, polyurethanes, polyamides and/or  
polytetrahydrofuranes.



- 5
6. Coating and adhesive material according to one or more of the preceding Claims, characterized in that the amount of said polymers in the first adhesive component, said polymers being reactive particularly with respect to isocyanates, is 20 to 100 wt.%, particularly 50 to 95 wt.%.  
10
7. Coating and adhesive material according to one or more of the preceding Claims, characterized in that one or more adhesive components comprise at least one resin.  
15
8. Coating and adhesive material according to Claim 7, characterized in that said resins are selected from the group consisting of aliphatic, cyclic or cycloaliphatic hydrocarbon resins, terpene phenol resins, cumarone-indene resins,  $\alpha$ -methyl styrene resins, polymerized tall resin ester or ketone aldehyde resins.  
20
9. Coating and adhesive material according to one or more of Claims 7 to 9, characterized in that the respective amount of said resin in one or more adhesive components is 0 to 70 wt.%.  
25
10. Coating and adhesive material according to one or more of the preceding Claims, characterized in that the first adhesive component comprises a non-reactive polymer.  
30
11. Coating and adhesive material according to Claim 10, characterized in that said non-reactive polymers are selected from the group consisting of ethylene/vinylacetate copolymers and/or polyolefins.  
35
12. Coating and adhesive material according to Claim 10 or 11, characterized in that said ethylene/vinylacetate copolymer has a vinylacetate content of between 12 and 40 %, particularly between 18 and 28 %, and a melting index of between 8 and 800, particularly between 150 and 500.
13. Coating and adhesive material according to any of Claims 10 to 12, characterized in that said polyolefins have average molecular weights  $M_n$  of between 5,000 and 25,000 g/mol, particularly between 10,000 and 20,000

g/mol, and softening ranges of between 80 and 170 °C, particularly between 80 and 130 °C.

- 5
14. Isocyanate-terminated coating and adhesive material according to one or more of the preceding Claims, characterized in that said reactive cross-linking component comprises an isocyanate solid at room temperature.
- 10
15. Coating and adhesive material according to Claim 14, characterized in that said solid isocyanates are selected from the group consisting of 4,4'-diisocyanato-diphenylmethane (MDI), 4,4',4''-tri-isocyanato-triphenylmethane, tris(4-isocyanatophenyl)-thiophosphate, 1,5-diisocyanato-naphthalene (NDI) and/or isomers thereof, dimers (dimerisates) of 2,4-diisocyanato-toluene (TDI) and/or of 1-isocyanato-3-isocyanatomethyl-3,5,5-trimethyl cyclohexane (IPDI) and/or their hydration products and/or trimers (trimerisates) of 1-isocyanato-3-isocyanatomethyl-3,5,5-trimethyl cyclohexane (TPDI).
- 15
16. Silane-terminated coating and adhesive material according to one or more of the preceding claims, characterized in that said reactive cross-linking component comprises at least one silanized polymer solid at room temperature.
- 20
17. Coating and adhesive material according to one or more of the preceding Claims, characterized in that the mixing ratio of the first and the second adhesive component is between 20:1 and 1:20.
- 25

**Abstract:**

The present invention relates to a multicomponent, isocyanate-terminated or silane-functionalized coating and adhesive material which is continuously  
5 produced by mixing the individual components and by heating the latter and continuing mixing until said components reach a liquid state, wherein the resulting reactive coating and adhesive material is fed for direct use especially in profile sheathing plants, coating plants or the like. In the case of isocyanate-terminated material, one component has a reactive high-molecular weight  
10 starting polymer and a second component has a reactive isocyanate-terminated cross-linking agent. In the case of the silane-functionalized material, one component has a high-molecular weight starting material and a second component has a reactive, silane-functionalized cross-linking agent.

5

10

**Translation of**

**Amended Claims of 7 March 2001**

15

**filed during International Preliminary Examination and  
on which the International Preliminary Search Report is based**

**Amended Claims:**

- 5
1. Reactive isocyanate-terminated multicomponent coating and/or adhesive material, said material comprising:
- (a) as a first component: an isocyanate-reactive starting polymer having a molecular mass of more than 8,000 g/mol; and
- 10 (b) as a second component: a reactive isocyanate-terminated cross-linking agent,
- wherein said material further comprises a polymer which is not isocyanate-reactive and which is selected from the group consisting of ethylene/vinyl-acetate copolymers and/or polyolefins.
- 15
2. The multicomponent coating and/or adhesive material of Claim 1, characterized in that said ethylene/vinylacetate copolymers have a vinyl-acetate content between 12 and 40 %, particularly 18 and 28 %, and melting indices between 8 and 800, particularly 150 and 500.
- 20
3. The multicomponent coating and/or adhesive material of Claim 1, characterized in that said polyolefins have average molecular weights  $M_n$  between 5,000 and 25,000 g/mol, particularly 10,000 and 20,000 g/mol, and softening ranges between 80 and 170 °C, particularly 80 and 130 °C.
- 25
4. The multicomponent coating and/or adhesive material according to any of the preceding Claims, characterized in that the content of said non-isocyanate-reactive polymer in said first component is 5 to 35 wt.%.
- 30
5. The multicomponent coating and/or adhesive material according to any of the preceding Claims, characterized in that said reactive cross-linking component comprises an isocyanate solid at room temperature, particularly wherein said isocyanate solid at room temperature is selected from the group consisting of 4,4'-diisocyanato-diphenylmethane (MDI),
- 35 4,4',4''-triisocyanato-triphenylmethane, tris-(4-isocyanatophenyl)-thiophosphate, 1,5-diisocyanato-naphthalene (NDI) and/or isomers thereof, dimers (dimerisates) of 2,4-diisocyanato-toluene (TDI) and/or of 1-

isocyanato-3-isocyanatomethyl-3,5,5-trimethylcyclohexane (IPDI) and/or their hydration products and/or trimers (trimerisates) of 1-isocyanato-3-isocyanatomethyl-3,5,5-trimethylcyclohexane (IPDI).

- 5      6.    The multicomponent coating and/or adhesive material according to any of the preceding Claims, characterized in that said isocyanate-reactive starting polymer has an average molecular weight between 8000 and 50,000 g/mol, particularly between 10,000 and 30,000 g/mol, particularly wherein said isocyanate-reactive starting polymer is preferably
- 10      selected from the group consisting of polyesters, polycaprolactonepolyesters, polyethers, polyurethanes, polyamides and polytetrahydrofuranes and/or has preferably at least two reactive groups with reactive hydrogen atoms per molecule.
- 15      7.    The multicomponent coating and/or adhesive material according to any of the preceding Claims, characterized in that the content of said isocyanate-reactive starting polymer in said first component is 20 to 100 wt.%, particularly 50 to 95 wt.%.
- 20      8.    The multicomponent coating and/or adhesive material according to any of the preceding Claims, characterized in that at least one adhesive component is present as a granulate (granular material).
- 25      9.    The multicomponent coating and/or adhesive material according to any of the preceding Claims, characterized in that one or more of the components comprise at least one resin, particularly wherein said resin is selected from the group consisting of aliphatic, cyclic or cycloaliphatic hydrocarbon resins, terpene phenol resins, cumarone-indene resins,  $\alpha$ -methyl styrene resins, polymerized tall resin
- 30      esters and ketone aldehyde resins and/or particularly wherein said resin is a resin having a low acid number with values less than 1 mg KOH/g and/or particularly wherein the respective content (amount) of said resin in one or more components is 0 to 70 wt.%, preferably 5 to 35 wt.%.
- 35

10. The multicomponent coating and/or adhesive material according to any of the preceding Claims, characterized in that the mixing ratio (blending ratio) of said first and said second component is between 20:1 and 1:20.
- 5 11. The multicomponent coating and/or adhesive material according to any of the preceding Claims, characterized in that at least one of said components comprises at least one further additive known per se with respect to reactive adhesives, particularly softeners (plasticizers) such as softeners (plasticizers) based on phthalic acids or phosphoric acid esters, glycol acetate, 10 high-boiling organic oils, esters or other additives inducing plastification, stabilizers, antioxidant agents (corrosion inhibitors), acid trappers (acid-trapping agents), and/or age inhibitors.
12. Reactive isocyanate-terminated multicomponent coating and/or adhesive material, said material comprising an isocyanate-reactive starting polymer having a molar mass of more than 8,000 g/mol; a reactive isocyanate-terminated cross-linking agent; and a polymer which is not isocyanate-reactive and selected from the group consisting of ethylene/vinylacetate copolymers and/or polyolefins. 15
13. The multicomponent coating and/or adhesive material of Claim 12, characterized in that it is moisture-reactive. 20
14. Method for preparing a reactive isocyanate-terminated multicomponent coating and/or adhesive material, particularly according to Claims 1 to 13, 25
- characterized in**
- that said reactive multicomponent coating and/or adhesive material is continuously produced by mixing (blending) the individual components and by their heating, under continued mixing, to a liquid state, 30
- wherein:
- 35 (a) a first component comprises an isocyanate-reactive starting polymer having a molar mass of more than 8,000 g/mol and

(b) a second component comprises a reactive isocyanate-terminated cross-linking agent and

wherein said multicomponent coating and/or adhesive material further comprises a polymer which is not isocyanate-reactive and which is selected from the group consisting of ethylene/vinylacetate copolymers and/or polyolefins,

and that the resulting reactive multicomponent coating and/or adhesive material, immediately after being prepared, is fed or conveyed, optionally via intermediate containers (buffer tanks), to a profile sheathing plant or to a coating plant for use.

15. Method for preparing a reactive isocyanate-terminated multicomponent coating and/or adhesive material, particularly according to Claims 1 to 13, **characterized in**

that said reactive multicomponent coating and/or adhesive material is continuously produced by mixing (blending) the individual components and by their heating, under continued mixing, to a liquid state,

wherein:

(a) a first component comprises an isocyanate-reactive starting polymer having a molar mass of more than 8,000 g/mol and

(b) a second component comprises a reactive isocyanate-terminated cross-linking agent and

wherein said multicomponent coating and/or adhesive material further comprises a polymer which is not isocyanate-reactive and which is selected from the group consisting of ethylene/vinylacetate copolymers and/or polyolefins,

and that the resulting reactive multicomponent coating and/or adhesive material, immediately after being prepared, is applied, optionally via intermediate containers (buffer tanks), by spraying, by injection, by nozzle (die) application or by roller application (roller coating).



16. The method according to any of the preceding claims, characterized in that the content of said non-isocyanate-reactive polymer in said first component preferably is in the range of from 5 to 35 wt%.
- 5 17. The method according to any of the preceding claims, characterized in that said ethylene/vinylacetate copolymers have a vinylacetate content between 12 and 40 %, particularly 18 and 28 %, and melting indices between 8 and 800, particularly 150 and 500.
- 10 18. The method according to any of the preceding claims, characterized in that said polyolefins have average molecular weights  $M_n$  between 5,000 and 25,000 g/mol, particularly 10,000 and 20,000 g/mol and softening ranges between 80 and 170 °C, particularly 80 and 130 °C.
- 15 19. The method according to any of the preceding claims characterized in that said reactive cross-linking component comprises an isocyanate which is solid at room temperature and/or particularly selected from the group consisting of 4,4'-diisocyanato-diphenylmethane (MDI), 4,4',4''-triisocyanato-triphenylmethane, tris-(4-isocyanatophenyl)-thiophosphate, 1,5-diisocyanato-naphthalene (NDI) and/or isomers thereof, dimers (dimerisates) of 2,4-diisocyanato-toluene (TDI) and/or of 1-isocyanato-3-isocyanatomethyl-3,5,5-trimethylcyclohexane (IPDI) and/or their hydration products and/or trimers (trimerisates) of 1-isocyanato-3-isocyanatomethyl-3,5,5-trimethylcyclohexane (IPDI).
- 20
- 25 20. The method according to any of the preceding claims characterized in that said isocyanate-reactive starting polymer has an average molecular weight between 8,000 and 50,000 g/mol, particularly 10,000 and 30,000 g/mol, particularly wherein said isocyanate-reactive starting polymer is preferably selected from the group consisting of polyesters, polycaprolactonepolyesters, polyethers, polyurethanes, polyamides and polytetrahydrofuranes and/or preferably has at least two reactive groups with reactive hydrogen atoms per molecule.
- 30

21. The method according to any of the preceding claims characterized in that the content of said isocyanate-reactive starting polymer in said first component is 20 to 100 wt.%, particularly 50 to 95 wt.%.
- 5 22. Method according to any of the preceding claims characterized in that at least one adhesive component is present as a granulate (granular material).
23. The method according to any of the preceding claims characterized in that one or more of the components have at least one resin,  
10 particularly wherein said resin is preferably selected from the group consisting of aliphatic, cyclic or cycloaliphatic hydrocarbon resins, terpene phenol resins, cumarone-indene resins,  $\alpha$ -methyl styrene resins, polymerized tall resin esters and/or the ketone aldehyde resins and/or particularly wherein said resin is preferably a resin with a low acid number  
15 with values lower than 1 mg KOH/g and/or particularly wherein the respective amount of said resin in one or more components is 0 to 70 wt.%, preferably 5 to 35 wt.%.
24. The method according to any of the preceding claims, characterized in that  
20 a mixing ratio (blending ratio) of said first and said second component between 20:1 and 1:20.
25. Use of the reactive isocyanate-terminated multicomponent coating and/or adhesive material according to Claims 1 to 13 for the bonding particularly  
25 of foam materials and cushions, upholstered furniture and mattresses, or for continuous coating such as profile sheathing (profile cladding), wherein the application is particularly performed by spraying, by injection, by nozzle (die) application or by roller application (roller coating).

**COMBINED DECLARATION FOR PATENT APPLICATION AND POWER OF ATTORNEY**  
Includes Reference to PCT International Applications

Attorney's Docket No.  
**4080-38PUS**

As a below named inventor, I hereby declare that:

My residence, post office address and citizenship are as stated below next to my name.

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled:

**MULTICOMPONENT COATING AND ADHESIVE MATERIAL**

the specification of which (check only one item below)

☐ is attached hereto

☐ was filed as United States application

Serial No. \_

on \_

and was amended

on \_ (if applicable).

☐ was filed as PCT international application

Number PCT/DE00/00384

on February 10, 2000

and was amended under PCT Article 19

on \_ (if applicable).

I hereby state that I have reviewed and understand the contents of the above-identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose information which is material to the patentability of the application in accordance with Title 37, Code of Federal Regulations, §1.56(a).

I hereby claim foreign priority benefits under Title 35, United States Code, §119 of any foreign application(s) for patent or inventor's certificate or of any PCT international application(s) designating at least one country other than the United States of America listed below and have also identified below any foreign application(s) for patent or inventor's certificate or any PCT international application(s) designating at least one country other than the United States of America filed by me on the same subject matter having a filing date before that of the application(s) of which priority is claimed.

**PRIOR FOREIGN/PCT APPLICATIONS AND ANY PRIORITY CLAIMS UNDER 35 U.S.C. 119:**

Country (if PCT, indicate "PCT")	Application Number	Date of Filing (day, month, year)	Priority Claimed Under 35 U.S.C. 119	
Germany	199 05 907.1	February 11, 1999	<input checked="" type="checkbox"/> YES	<input type="checkbox"/> NO
PCT	PCT/DE00/00384	February 10, 2000	<input checked="" type="checkbox"/> YES	<input type="checkbox"/> NO
			<input type="checkbox"/> YES	<input type="checkbox"/> NO
			<input type="checkbox"/> YES	<input type="checkbox"/> NO
			<input type="checkbox"/> YES	<input type="checkbox"/> NO
			<input type="checkbox"/> YES	<input type="checkbox"/> NO
			<input type="checkbox"/> YES	<input type="checkbox"/> NO

Combined Declaration for Patent Application and Power of Attorney (Continued)  
(Includes Reference to PCT International Applications)

Attorney's Docket No.  
4080-38PUS

I hereby claim the benefit under Title 35, United States Code, §120 of any United States application(s) or PCT international application(s) designating the United States of America that is/are listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in that/those prior application(s) in the manner provided by the first paragraph of Title 35, United States Code, §112, I acknowledge the duty to disclose material information as defined in Title 37, Code of Federal Regulations, §1.56(a) which occurred between the filing date of the prior application(s) and the national or PCT international filing date of this application:

PRIOR U.S. APPLICATIONS OR PCT INTERNATIONAL APPLICATIONS DESIGNATING THE U.S. FOR BENEFIT UNDER 35 U.S.C. 120:

U.S. APPLICATIONS		STATUS (check one)		
U.S. APPLICATION NUMBER	U.S. FILING DATE	PATENTED	PENDING	ABANDONED
PCT APPLICATIONS DESIGNATING THE U.S.				
PCT APPLICATION NO.	PCT FILING DATE	U.S. SERIAL NUMBERS ASSIGNED (if any)		
PCT/DE00/00384	February 10, 2000		X	

**POWER OF ATTORNEY:** As a named inventor, I hereby appoint the following attorney(s) and/or agent(s) to prosecute this application and transact all business in the Patent and Trademark Office connected therewith (*List name and registration number*)

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Combined Declaration for Patent Application and Power of Attorney (Continued) (Includes Reference to PCT International Applications)				Attorney's Docket No. 4080-38PUS
2 0 3	FULL NAME OF INVENTOR	FAMILY NAME	FIRST GIVEN NAME	SECOND GIVEN NAME
	RESIDENCE, CITIZENSHIP	CITY	STATE OR FOREIGN COUNTRY	COUNTRY OF CITIZENSHIP
	POST OFFICE ADDRESS	POST OFFICE ADDRESS	CITY	STATE & ZIP CODE/COUNTRY
<p>I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under §1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.</p>				
SIGNATURE OF INVENTOR 201 <i>[Signature]</i>		SIGNATURE OF INVENTOR 202 <i>[Signature]</i>		SIGNATURE OF INVENTOR 203
DATE 13.9.01		DATE 14.9.01		DATE